<Division of Environmental Chemistry>Molecular Materials Chemistry

Division of Environmental Chemistry
– Molecular Materials Chemistry –

http://molmat.kuicr.kyoto-u.ac.jp/index-e.html

Prof
KAJI, Hironori
(D Eng)

Assist Prof
SHIZU, Katsuyuki
(D Eng)

Assist Prof
SUZUKI, Katsuaki
(D Human & Envirnmtl. Studies)

Techn Staff
OHMINE, Kyoko

Techn Staff
MAENO, Ayaka

Assist Techn Staff

YAMADA, Kyoko

Researcher

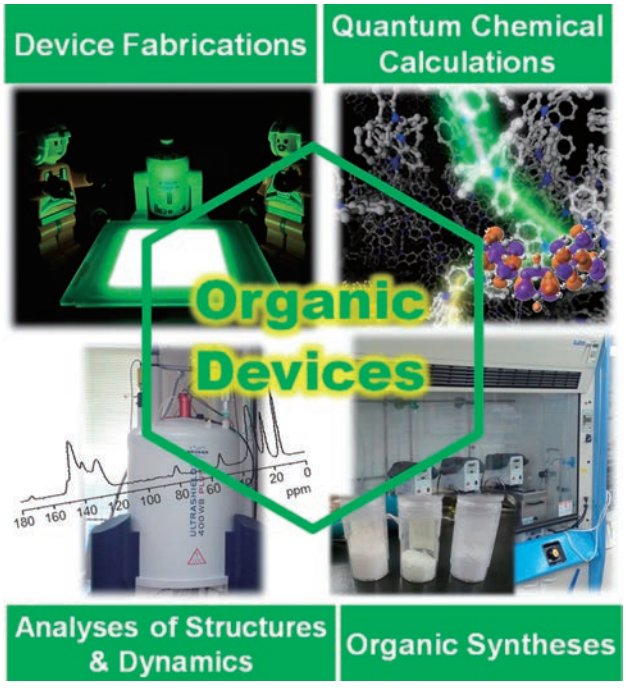
NAKAGAWA, Hiromichi (D Eng)

Students

KUBO, Shosei (D3)
WADA, Yoshimasa (D2)
WAKAYAMA, Rin (M2)
WAKISAKA, Yasuaki (M2)
SAWADA, Ayaka (M1)
MISONO, Tomoya (M1)
HU, Weizhe (RS)
MORIWAKI, Kazuki (UG)
SAKAI, Kahori (UG)

Scope of Research

Our research goal is to develop high-performance organic electroluminescence devices, organic solar cells, and polymer materials. Toward this, we carry out syntheses, device fabrications, precise structure characterizations, and quantum chemical calculations for high functional organic materials. Along with exploring novel synthetic routes and novel devices, we perform detailed analyses of structures and dynamics, mainly by sophisticated solid-state NMR spectroscopy, in order to obtain structure–dynamics–property relationships.



KEYWORDS

Organic Light-Emitting Diodes
Organic Solar Cell
Solid-State NMR
Quantum Chemical Calculation
Amorphous Materials

Selected Publications

Kubo, S.; Kaji, H., Parameter-Free Multiscale Simulation Realising Quantitative Prediction of Hole and Electron Mobilities in Organic Amorphous System with Multiple Frontier Orbitals, *Sci. Rep.*, **8,** [5203-1]-[5203-9] (2018).

Sasabe, H.; Sato, R.; Suzuki, K.; Watanabe, Y.; Adachi, C.; Kaji, H.; Kido, J., Ultrahigh Power Efficiency Thermally Activated Delayed Fluorescent OLEDs by the Strategic Use of Electron-Transport Materials, *Adv. Optical Mater.*, **6,** [1800376-1]-[1800376-5] (2018).

Suzuki, F.; Kubo, S.; Fukushima, T.; Kaji, H., Effects of Structural and Energetic Disorders on Charge Transports in Crystal and Amorphous Organic Layers, *Sci. Rep.*, **8,** [5203-1]-[5203-9] (2018).

Wada, Y.; Kubo, S.; Kaji, H., Adamantyl Substitution Strategy for Realizing Solution-Processable Thermally Stable Deep-Blue Thermally Activated Delayed Fluorescence Materials, *Adv. Mater.*, **30,** [1705641-1]-[1705641-8] (2018).

Adamantyl Substitution Strategy for Realizing Solution-Processable Thermally Stable Deep-Blue Thermally Activated Delayed Fluorescence Materials

Highly efficient solution-processable emitters, especially deep-blue emitters, are greatly desired to develop low-cost and low-energy-consumption organic light-emitting diodes (OLEDs). A recently developed class of potentially metal-free emitters, thermally activated delayed fluorescence (TADF) materials, are promising candidates, but solution-processable TADF materials with efficient blue emissions are not well investigated. In this study, first the requirements for the design of efficient deep-blue TADF materials are clarified, on the basis of which, adamantyl-substituted TADF molecules are developed. The substitution not only endows high solubility and excellent thermal stability but also has a critical impact on the molecular orbitals, by pushing up the lowest unoccupied molecular orbital energy and triplet energy of the molecules. In the application to OLEDs, an external quantum efficiency (EQE) of 22.1% with blue emission having Commission Internationale de l'Eclairage (CIE) coordinates of (0.15, 0.19) is realized. A much deeper blue emission with CIE (0.15, 0.13) is also achieved, with an EQE of 11.2%. These efficiencies are the best yet among solution-processed TADF OLEDs of CIE $y < 0.20$ and $y < 0.15$, as far as known. This work demonstrates the validity of adamantyl substitution and paves a pathway for straightforward realization of solution-processable efficient deep-blue TADF emitters.

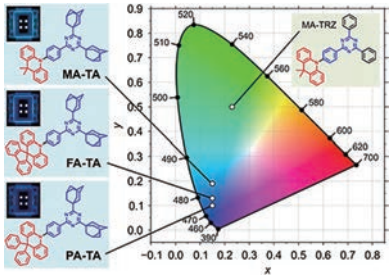


Figure 1. Molecular structures of MA-TA, FA-TA, and PA-TA, and their electroluminescence properties.

Effects of Structural and Energetic Disorders on Charge Transports in Crystal and Amorphous Organic Layers

Understanding charge transports in organic films is important for both fundamental science and practical applications. Here, contributions of off-diagonal (structural) and diagonal (energetic) disorders to charge transports were clarified using molecular-based multiscale simulation. These disorders, important for understanding charge transport in organic systems, are investigated by comparing crystal and amorphous aggregates of N,N'-diphenyl-N,N'-bis(1-naphthyl)-1,1'-biphenyl-4,4'-diamine (NPD). Although NPD has been used as a hole transport material, it also exhibits comparable electron mobility experimentally. The experimental mobility and its electric field dependence in amorphous layers were reasonably reproduced by the multiscale simulation, confirming the electron transport properties of NPD. We assumed that the structural disorder would lower mobilities; however, the mobilities were found to be independent of the degree of structural disorder. Energetic disorder markedly lowered charge mobility instead. Charge migration in crystals was dominated by maximum electronic coupling pairs, whereas small electronic coupling pairs significantly contributed to charge transport in amorphous aggregate.

Parameter-Free Multiscale Simulation Realising Quantitative Prediction of Hole and Electron Mobilities in Organic Amorphous System with Multiple Frontier Orbitals

In amorphous organic semiconducting systems, hole and electron transfer has been considered to occur based on the overlap of highest occupied molecular orbitals (HOMOs) and that of lowest unoccupied molecular orbitals (LUMOs) between two adjacent molecules, respectively. Other molecular orbitals (MOs), HOMO−1, HOMO−2, ... and LUMO+1, LUMO+2, ..., have been neglected in charge transport calculations. However, these MOs could potentially contribute to charge transport. In this study, our multiscale simulations show that carriers are effectively transported not only via HOMOs or LUMOs but also via other MOs when the MOs are close in energy. Because these multiple MOs are active in charge transports, here we call them multiple frontier orbitals. Molecules with multiple frontier orbitals are found to possess high carrier mobility. The findings in this study provide guidelines to aid design of materials with excellent charge transport properties.

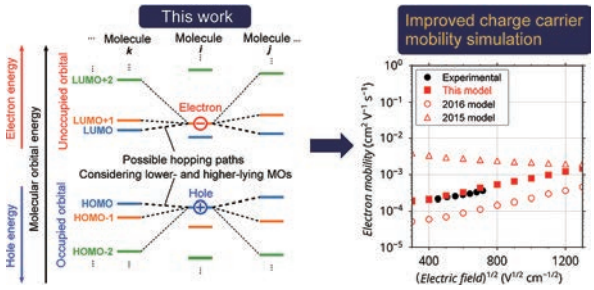


Figure 2. The charge transport simulation model for this work and electric field dependence of electron mobility.